PATENT
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Richard F. Lemuth

Date: July 24, 2006

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re the application of	
MAREK MATUSZ ET AL	
Serial No. 10/816,543	Group Art Unit: 1725
Filed April 1, 2004	Examiner: Christina Ann Johnson
SILVER-CONTAINING CATALYSTS, THE MANUFACTURE OF SUCH SILVER-CONTAINING CATALYSTS, AND THE USE THEREOF	

COMMISSIONER FOR PATENTS
P. O. Box 1450
Alexandria, VA 22313-1450

Sir:

DECLARATION UNDER 37 C.F.R. 1.132

The undersigned, Dr. Paul M. McAllister, citizen of the United States of America and residing in Houston, TX, declare and say that:

1. I am a 1987 graduate of the University of Kansas with a Bachelor of Science degree in Chemical Engineering; a 1989 graduate of Notre Dame University with a Master of Sciences degree in Chemical Engineering; and a 1991 graduate of Notre Dame University with a Doctor of Philosophy Degree in Chemical Engineering.

2. In 1991, I joined Shell Oil Company as an Associate Research Engineer in the Environmental Research and Development Department at Shell's Westhollow Research Center. I was involved in the modeling of transport phenomena in porous media and remediation of soil and groundwater.

3. In 1998, I was transferred to the Ethylene Oxide ("EO") Catalyst Department. My activities for Shell have involved technical support for EO producers, modeling of EO catalyst performance, scale up of novel catalysts and carriers from EO catalyst pilot plant to EO catalyst commercial unit, and oversight of the EO catalyst testing facilities.

4. I am currently an inventor on three pending patent applications relating to ethylene oxide processes.

5. I have read the above-identified patent application, the PTO Office Actions dated 08/09/05 and 01/25/06, the Applicant's Response dated 11/14/05, and the prior art cited by the Examiner (Johnson US 4,532,231 and Liu et al. US 6,511,938).

6. As noted in the subject patent application, Catalyst Examples C, D and E are according to the invention, and Catalyst Examples A and B are comparisons. These catalysts were prepared using hollow cylinder shaped carrier pellets having an outside diameter and an internal (bore) diameter. It is understood that the terms "bore size", "bore diameter", and "internal diameter", as used throughout, are intended to be synonymous. As shown in Table IV, Catalysts C, D and E have an unexpected balance of property advantages over Catalysts A and B, in that Catalysts C, D and E have higher activity and/or selectivity, as shown by lower temperatures at equivalent time periods, along with good and acceptable pressure drop in the reactor. This performance was not expected, since it had been predicted by a commonly accepted scientific correlation (the Ergun Correlation) that catalysts with such smaller internal diameters would have much larger pressure drops in the reactor compared to catalysts with larger internal diameters.

7. In order to test the effect of smaller internal diameters, I conducted various experiments with hollow EO catalyst carrier shapes of varying size to determine the resulting pressure drops when loaded into a test vessel. In one set of experiments, I compared nominal 8 mm outside diameter ("OD") carrier pellets having actual average bore sizes of 3.27, 2.90, 2.70, 1.96, 1.30, and 0.78 mm along with a carrier with no hole (internal diameter defined as 0 mm), against a nominal 8 mm OD carrier pellet having a standard actual average bore size of 3.2 mm. The nominal 8 mm OD carrier pellets had actual average outside diameters ranging from 8.4 to 8.7 mm. After each carrier sample was loaded in the test vessel, the pressure drop was measured with nitrogen flow over the range of Reynolds numbers (Re_{tube} based on tube ID) typical for commercial EO reactors. Re_{tube} was varied from 7500 to 35000. The commonly used Ergun Correlation (EC) was used in the evaluation of the pressure drop data. This Ergun Correlation is shown below:

$$\frac{\Delta P}{L} = C_1 \frac{\mu V_o}{D_p^{\,2}} \frac{(1-\varepsilon)^2}{\varepsilon^3} + C_2 \frac{\rho V_o^2}{D_p} \frac{(1-\varepsilon)}{\varepsilon^3} \qquad \text{(EC)}$$

Where,

$\frac{\Delta P}{L}$ is the pressure drop per unit reactor length,

V_o is the superficial gas velocity,

μ is the gas viscosity,

ε is the void fraction of the packed bed external to the pellets,

ρ is the gas density,

D_p is the effective particle diameter,

C_1 is the laminar term coefficient,

and

C_2 is the turbulent term coefficient.

The effective particle diameter can be defined as:

$$\boldsymbol{D_p} = 6\frac{\boldsymbol{V_{solid}}}{\boldsymbol{A_{exterior}}} \qquad (\boldsymbol{D_p})$$

Where,

V_{solid} is the geometric solid volume of each particle,

and

$A_{exterior}$ is the geometric external surface area of a particle.

For the high Re_{tube} conditions used in this testing and typical for EO reactors, the first term of Equation (EC), which is proportional to the velocity, is small relative to the second term, which is proportional to the velocity squared. Thus, it was found that the first term could be neglected, and the measured pressure drops were proportional to the product of the average gas density and the average superficial velocity squared. This turned out to be true for all conditions and carrier samples tested.

8. Since all parameters in the Ergun Correlation (EC) can be calculated from the hollow cylinder characterization information, comparisons were made with predicted pressure drops. Figure 1 shows the predicted pressure drop change as calculated by the Ergun Correlation compared to the actual measurements for decreasing bore size with 8 mm OD cylinders, using the standard 3.2 mm bore 8 mm OD cylinder as the reference. The actual average outside diameters and bore sizes were utilized in Figure 1. Using the Ergun Correlation, I expected the pressure drop to increase by **60-70%** as the bore size was reduced from 3.2 mm to 0 mm with 8 mm OD cylinders. In reality, the pressure drop increase was **only 20%** for the 8 mm OD solid cylinder. This was an **unexpected** finding.

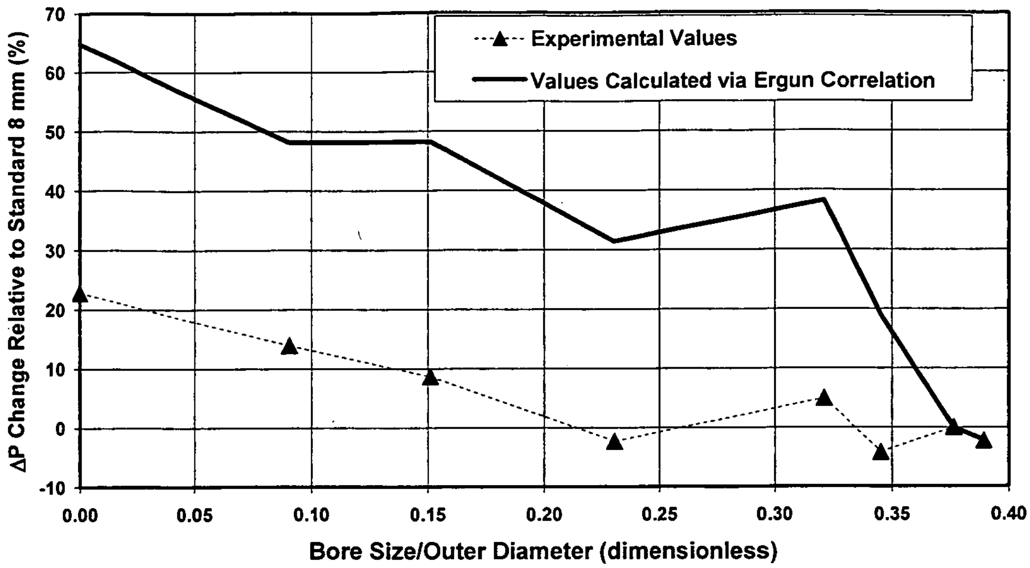

Figure 1. Comparison of *a priori* Ergun correlation predictions and actual ΔP changes for 8 mm OD cylinders with varied bore sizes relative to the standard 3.2 mm bore 8 mm OD cylinder (bore size/outer diameter ratio of 0.38).

9. Significant deviations between the Ergun Correlation predictions and experimental data occurred with relatively small changes in the internal diameter to outside diameter ("ID/OD") ratio of the pellets. For example, reducing the ID/OD ratio from 0.38 to 0.34 was predicted to increase pressure drop by 19%. In reality, the pressure drop was unchanged or slightly lower (-4%).

10. While the size and shape of the support for EO catalysts are variable factors, prior to the subject patent application, it would have been predicted by the Ergun Correlation that carrier shapes with such small internal diameters would not be acceptable for commercial EO production. Therefore, the invention claimed in the subject patent application is unexpected, and would not be expected by any reasonable reading of the Johnson and Liu et al. prior art.

11. I declare further that all statements made herein of my knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Respectfully submitted,

Paul M. McAllister

This 26 day of June, 2006